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EUROPEAN PATENT APPLICATION

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(54) **Catalyst for polyester production, process for producing polyester using the catalyst, polyester obtained by the process, and uses of the polyester**

(57) The present invention provides a catalyst for polyester production capable of producing a polyester with high catalytic activity and a process for producing a polyester using the catalyst. The catalyst for polyester production comprises a solid titanium compound which is obtained by dehydro-drying a hydrolyzate obtained by hydrolysis of a titanium halide and which has a molar ratio (OH/Ti) of a hydroxyl group (OH) to titanium (Ti) exceeding 0.09 and less than 4. The present invention also provides a method to obtain a polyester having a small increase of the acetaldehyde content during the molding.

This method comprises bringing a polyester, which is obtained by the use of a titanium compound catalyst and in which the reaction has been completed, into contact with a phosphoric ester aqueous solution or the like having a concentration of not less than 10 ppm in terms of phosphorus atom. The present invention further provides a polyester having excellent transparency and tint and molded products of the polyester such as a blow molded article, a film, a sheet and a fiber. The polyester is obtained by polycondensing an aromatic dicarboxylic acid or an ester-forming derivative thereof and an

EP 1 013 692 A3

aliphatic diol or an ester-forming derivative thereof in the presence of a catalyst for polyester production which comprises a polycondensation catalyst component comprising a solid titanium compound and a co-catalyst com-

ponent comprising a magnesium compound. This polyester has a titanium content of 1 to 100 ppm, a magnesium content of 1 to 200 ppm and a weight ratio (Mg/Ti) of magnesium to titanium of not less than 0.01.

Fig.1

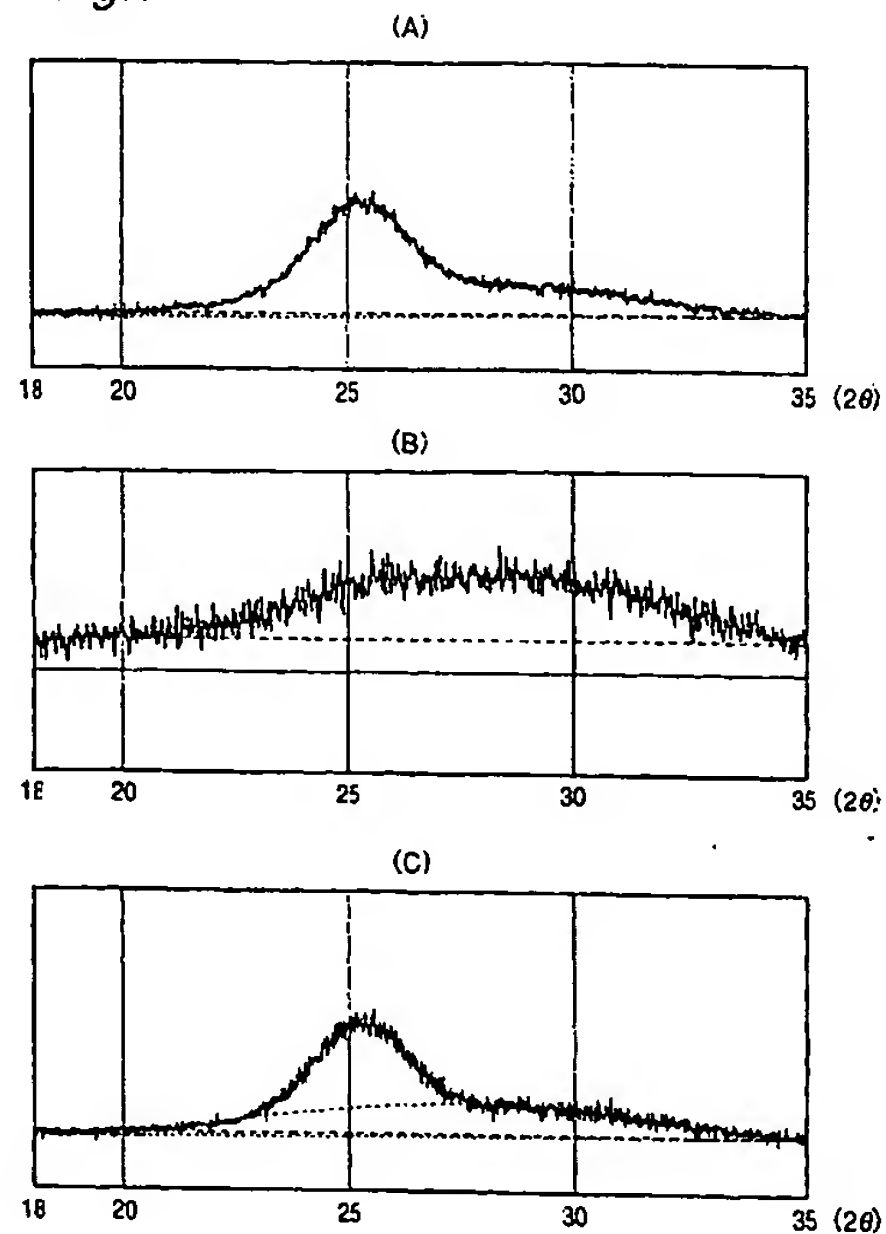
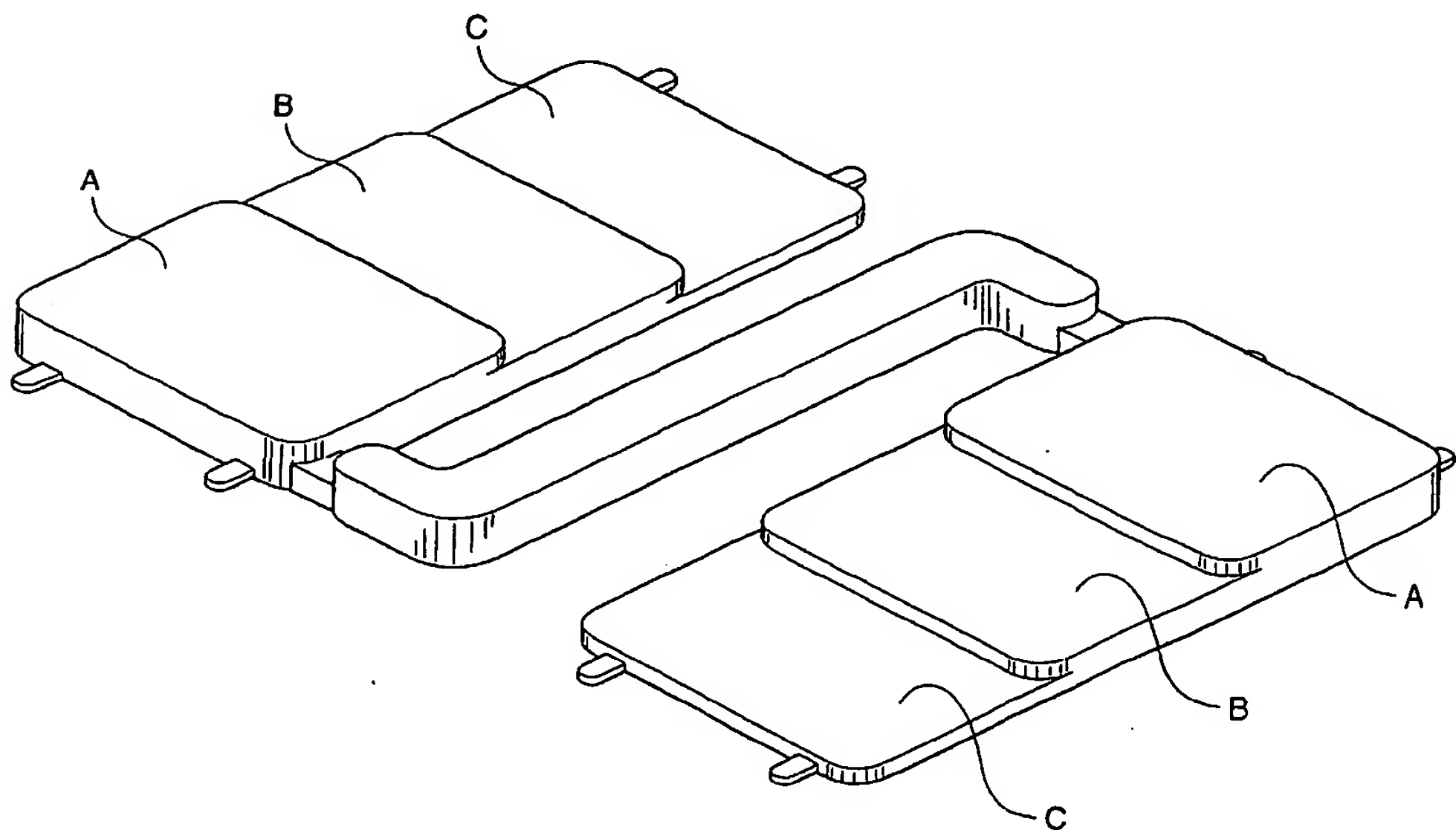


Fig.2



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PARTIAL EUROPEAN SEARCH REPORT

which under Rule 45 of the European Patent Convention shall be considered, for the purposes of subsequent proceedings, as the European search report

Application Number

EP 99 12 5906

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X	US 3 056 817 A (WERBER FRANK X ET AL) 2 October 1962 (1962-10-02) * column 2, line 24 - line 44 * * column 6, line 50 - line 60 * * examples I,VI,VII,IX * * claims 3,4 *	1,4, 11-13,31	
X	US 5 656 716 A (SCHMIDT WOLFGANG ET AL) 12 August 1997 (1997-08-12) * column 2, line 35 - line 43 * * column 4, line 8 - line 15 *	1-4,7, 11-13,31	
X	GB 1 179 356 A (TOA GOSEI CHEM IND CO. LTD.) 28 January 1970 (1970-01-28) * examples 1-8,15-20,26-34 *	1-4,11, 12, 15-18,31	
X	CH 455 290 A (INVENTA AG) 28 June 1968 (1968-06-28) * examples *	1,4, 11-14,31	C08G B01J C08L
-/--			
INCOMPLETE SEARCH			
<p>The Search Division considers that the present application, or one or more of its claims, does/do not comply with the EPC to such an extent that a meaningful search into the state of the art cannot be carried out, or can only be carried out partially, for these claims.</p> <p>Claims searched completely :</p> <p>Claims searched incompletely :</p> <p>Claims not searched :</p> <p>Reason for the limitation of the search:</p> <p style="text-align: center;">see sheet C</p>			
Place of search		Date of completion of the search	Examiner
MUNICH		13 April 2004	Russell, G
CATEGORY OF CITED DOCUMENTS		<p>T : theory or principle underlying the invention</p> <p>E : earlier patent document, but published on, or after the filing date</p> <p>D : document cited in the application</p> <p>L : document cited for other reasons</p> <p>8 : member of the same patent family, corresponding document</p>	
<p>X : particularly relevant if taken alone</p> <p>Y : particularly relevant if combined with another document of the same category</p> <p>A : technological background</p> <p>O : non-written disclosure</p> <p>P : intermediate document</p>			

EPO FORM 1503 03/02 (P04C07)



European Patent
Office

INCOMPLETE SEARCH SHEET C

Application Number

EP 99 12 5906

Claim(s) searched completely:
1-20, 23-58, 73-76

Claim(s) searched incompletely:
59-72, 77-79

Claim(s) not searched:
21, 22

Reason for the limitation of the search:

Present independent claims 59, 62, and 65 relate to polyesters and molded articles defined (inter alia) by reference to the following parameters:

P1: acetaldehyde content when measured after heat treatment at specific conditions (claim 59),

P2: cyclic trimer content when measured after heat treatment at specific conditions (claim 52),

P3: ratio of flow length to flow thickness in injection molding at temperature and intrinsic viscosity.

The use of these parameters in the present context is considered to lead to a lack of clarity within the meaning of Article 84 EPC. Said parameters do not appear to be standard in the art, such that they could not be found during search. As P1, P2, and/or P3 are not disclosed in prior documents, it is impossible to compare the parameters the applicant has chosen to employ with what is set out in the prior art to establish if these prior disclosures fall within the claims. For novelty, such features may be inherently present in prior polyesters (see EPO Guidelines C-III, 4.7a and C-IV, 7.5). The lack of clarity is such as to render a meaningful complete search impossible. Consequently, the search has been restricted to polyester and molded articles therefrom of the art regardless of these parameters.

Claim 65 relates, in addition, to a polyester defined by reference to a desirable characteristic or property, namely the desire to achieve a certain (L/T) ratio on injection molding at 290°C. The claim covers all products having this characteristic or property, whereas the application provides support within the meaning of Article 84 EPC and/or disclosure within the meaning of Article 83 EPC for only a very limited number of such products. In the present case, the claims so lack support, and the application so lacks disclosure, that a meaningful search over the whole of the claimed scope is impossible. Independent of the above reasoning, claim 65 also lacks clarity (Article 84 EPC). An attempt is made to define the polyester by reference to a result to be achieved without providing the technical features necessary to achieve this result. Again, this lack of clarity in the present case is such as to render a meaningful search over the whole of the claimed scope impossible.

As a result, claims 59-64 and 77-79 were searched partially based on polyesters containing titanium and metal atom M, and having a specific content of acetaldehyde or cyclic trimer, respectively. Claims 65 was partly searched based on the content of dependent claims 66 to 72, which pertain to methods of obtaining such a polyester, i.e., of achieving the desired characteristic, fulfilling the parameter P3. Thus, claims 59-72, and 77-79 could only be searched incompletely.



European Patent
Office

**INCOMPLETE SEARCH
SHEET C**

Application Number
EP 99 12 5906

Claims 21 and 22 relating to subject invention 3., for which no additional search fees were paid, were not searched.



European Patent
Office

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Application Number
EP 99 12 5906

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X	WO 98/56848 A (SEIDEL ULF ;MARTL MICHAEL GERD (DE); AKZO NOBEL NV (NL)) 17 December 1998 (1998-12-17) * examples 1-26 * * claims 1-3,7,16-19 *	23,25, 28-31, 34,35	
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X	DATABASE WPI Section Ch, Week 197415 Derwent Publications Ltd., London, GB; Class A23, AN 1974-28355V XP002276398	23,25,31	
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EPO FORM 1503 03.82 (P04C10)

P 702 - EP 10

EP 1 013 692 A3



European Patent
Office

PARTIAL EUROPEAN SEARCH REPORT

Application Number
EP 99 12 5906

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Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
X	DATABASE CA 'Online! CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; KIMURA, TOMIAKI ET AL: "Polyesters" retrieved from STN Database accession no. 83:115430 XP002276395	23,25,31	
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X	DATABASE CA 'Online! CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; TSUNAWAKI, KIYOKAZU ET AL: "Aromatic polyesters" retrieved from STN Database accession no. 82:4777 XP002276396	23,25,31	
A	* abstract * & JP 49 011754 B (TEIJIN LTD.) 19 March 1974 (1974-03-19) ---	26,28-30	
X	DATABASE CA 'Online! CHEMICAL ABSTRACTS SERVICE, COLUMBUS, OHIO, US; TSUJI, TAKAAKIRA ET AL: "Poly(tetramethylene terephthalate)" retrieved from STN Database accession no. 85:78958 XP002276397	23,25,31	TECHNICAL FIELDS SEARCHED (Int.Cl.7)
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EPO FORM 1503 03 82 (P04C10)



European Patent
Office

PARTIAL EUROPEAN SEARCH REPORT

Application Number

EP 99 12 5906

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Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
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A	US 4 374 949 A (MASSEY FRED L ET AL) 22 February 1983 (1983-02-22) * claim 1 *	39,58	
A	US 3 965 071 A (MCCLELLAND EUAN) 22 June 1976 (1976-06-22) * claim 1 *	28,29, 34-37, 42,50	TECHNICAL FIELDS SEARCHED (Int.Cl.7)
D,X	PATENT ABSTRACTS OF JAPAN vol. 017, no. 442 (C-1097), 16 August 1993 (1993-08-16) -& JP 05 097990 A (TEIJIN LTD), 20 April 1993 (1993-04-20)	42-44	
A	* abstract *	62,77	
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EPO FORM 1503 03.92 (P04C10)

PARTIAL EUROPEAN SEARCH REPORT

Application Number
EP 99 12 5906

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
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X	CH 655 938 A (INVENTA AG) 30 May 1986 (1986-05-30) * claims 1,3-5,7; table 1 *	45-49	
X	PATENT ABSTRACTS OF JAPAN vol. 1997, no. 02, 28 February 1997 (1997-02-28) -& JP 08 283393 A (MITSUI PETROCHEM IND LTD), 29 October 1996 (1996-10-29) * abstract * * paragraphs '0047!', '0048!', '0085!' *	56-79	TECHNICAL FIELDS SEARCHED (Int.Cl.7)
X	US 5 688 899 A (SUBLETT BOBBY JONES ET AL) 18 November 1997 (1997-11-18) * abstract * * column 3, line 40 - line 48; table II * * claims 1,2,6-10 *	56-73, 75,77	
X	US 4 128 533 A (KOHLER ARMIN ET AL) 5 December 1978 (1978-12-05) * claims *	56-73,76	
X	PATENT ABSTRACTS OF JAPAN vol. 004, no. 050 (C-007), 16 April 1980 (1980-04-16) -& JP 55 023136 A (TEIJIN LTD), 19 February 1980 (1980-02-19) * abstract *	56-74, 77-79	

Application Number

PARTIAL EUROPEAN SEARCH REPORT

EP 99 12 5906

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Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
X	US 5 852 164 A (FUJIMOTO MASAHIKO ET AL) 22 December 1998 (1998-12-22) * column 4, line 55 - line 67 * * column 10, line 5 - line 12 * * claims 1,2,5-10,12-16 *	58-70, 72-75, 77-79	TECHNICAL FIELDS SEARCHED (Int.Cl.7)
X	US 5 270 444 A (SAKAI MASAYUKI ET AL) 14 December 1993 (1993-12-14) * column 3, line 39 - column 4, line 29 * * column 13, line 5 - line 16 *	65-70, 72-75, 77-79	
A	US 3 663 600 A (MCCONNELL WAYNE V ET AL) 16 May 1972 (1972-05-16) * abstract *	50-55	



European Patent
Office

Application Number

EP 99 12 5906

CLAIMS INCURRING FEES

The present European patent application comprised at the time of filing more than ten claims.

- ☐ Only part of the claims have been paid within the prescribed time limit. The present European search report has been drawn up for the first ten claims and for those claims for which claims fees have been paid, namely claim(s):
- ☐ No claims fees have been paid within the prescribed time limit. The present European search report has been drawn up for the first ten claims.

LACK OF UNITY OF INVENTION

The Search Division considers that the present European patent application does not comply with the requirements of unity of invention and relates to several inventions or groups of inventions, namely:

see sheet B

- ☐ All further search fees have been paid within the fixed time limit. The present European search report has been drawn up for all claims.
- ☐ As all searchable claims could be searched without effort justifying an additional fee, the Search Division did not invite payment of any additional fee.
- ☒ Only part of the further search fees have been paid within the fixed time limit. The present European search report has been drawn up for those parts of the European patent application which relate to the inventions in respect of which search fees have been paid, namely claims:
1-20, 23-79
- ☐ None of the further search fees have been paid within the fixed time limit. The present European search report has been drawn up for those parts of the European patent application which relate to the invention first mentioned in the claims, namely claims:



European Patent
Office

LACK OF UNITY OF INVENTION
SHEET B

Application Number
EP 99 12 5906

The Search Division considers that the present European patent application does not comply with the requirements of unity of invention and relates to several inventions or groups of inventions, namely:

1. Claims: 1,4-6,11-14,31

Solid titanium compound catalyst suitable for polyester production obtained by dehydro-drying a hydrolyzate obtained from hydrolyzing a titanium halide, the catalyst having a molar ratio (OH/Ti) of >0.09 to < 4 ; catalyst system for polyester production comprising said catalyst and (II) a co-catalyst; processes for preparing said catalyst; and process for producing polyester using said catalyst.

2. Claims: 2-10,15-20,31

Solid titanium compound catalyst suitable for polyester production obtained by dehydro-drying a hydrolyzate obtained from hydrolyzing a mixture titanium halide and a compound or its precursor selected from at least one element other than titanium, the catalyst having a molar ratio (OH/Ti) of >0.09 to < 4 ; catalyst system for polyester production comprising said catalyst and (II) a co-catalyst; processes for preparing said catalyst; and process for producing polyester using said catalyst.

3. Claims: 21,22,31

Solid titanium compound catalyst for polyester production having a crystallinity, calculated from a X-ray diffraction pattern having 2θ of 18 to 35° , of not more than 50%, said catalyst obtained by dehydro-drying titanium hydroxide; catalyst system for polyester production comprising said titanium catalyst and (II) co-catalyst; and process for producing polyester using said catalyst.

4. Claims: 23-41

Catalyst for polyester production comprising a slurry obtained by heating a mixture of (A) a hydrolyzate obtained by hydrolyzing a titanium compound, or a hydrolyzate obtained by hydrolyzing a titanium compound and a compound or its precursor selected from at least one element other than titanium, (B) a basic compound, and (C) an aliphatic diol; said catalyst (A) comprising (D) a metallic phosphate; said catalyst system (A) comprising (F) at least one phosphorous compound, and (G) an aliphatic diol; a process for producing a polyester using said catalyst; a process for producing a polyester using said catalyst system with a (II) co-catalyst; a process for producing a polyester using said catalyst system with a phosphoric ester, and a (II) co-catalyst; a process for producing a polyester using said



European Patent
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LACK OF UNITY OF INVENTION
SHEET B

Application Number
EP 99 12 5906

The Search Division considers that the present European patent application does not comply with the requirements of unity of invention and relates to several inventions or groups of inventions, namely:

catalyst system with a compound selected from cyclic lactone compounds and hindered phenol compounds, and a (II) co-catalyst; a process for producing a polyester of increased molecular weight using said catalyst system, and a (II) co-catalyst, an a tint adjusting agent.

5. Claims: 42-55

Methods of treating of polyester prepared using a titanium compound catalyst comprising bringing a polyester in contact with a phosphorous acid aq. solution; bringing a polyester in contacting with an organic solvent; or bringing a polyester in contact with an organic solvent solution of phosphorous compound.

6. Claims: 56-64, 73-77

Polyesters (P-1 to P-4) characterized in terms of titanium atom content, specific metal atom content, tint adjusting agent content, acetaldehyde content, and cyclic trimer content.

7. Claims: 65-72, 78, 79

Polyester (P-5) characterized in terms of a relation between injection molding features and intrinsic viscosity of the molded polyester; in terms of catalyst used for its production; its titanium atom and magnesium atom contents; preform; and blow-molded article made therefrom.

ANNEX TO THE EUROPEAN SEARCH REPORT ON EUROPEAN PATENT APPLICATION NO.

EP 99 12 5906

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EPO FORM P0459

For more details about this annex : see Official Journal of the European Patent Office, No. 12/82

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EP 99 12 5906

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EPO FORM P0459

For more details about this annex : see Official Journal of the European Patent Office, No. 12/82

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EP 99 12 5906

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